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Integrated lignocellulosic value chains in a growing bioeconomy: Status quo and perspectives

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Abstract

Lignocellulose is the most abundant biomass on Earth, with an estimated 181.5 billion tonnes produced annually. Of the 8.2 billion tonnes that are currently used, about 7 billion tonnes are produced from dedicated agricultural, grass and forest land and another 1.2 billion tonnes stem from agricultural residues. Economic and environmentally efficient pathways for production and utilization of lignocellulose for chemical products and energy are needed to expand the bioeconomy. This opinion paper arose from the research network “Lignocellulose as new resource platform for novel materials and products” funded by the German federal state of Baden-Württemberg and summarizes original research presented in this special issue. It first discusses how the supply of lignocellulosic biomass can be organized sustainably and suggests that perennial biomass crops (PBC) are likely to play an important role in future regional biomass supply to European lignocellulosic biorefineries. Dedicated PBC production has the advantage of delivering biomass with reliable quantity and quality. The tailoring of PBC quality through crop breeding and management can support the integration of lignocellulosic value chains. Two biorefinery concepts using lignocellulosic biomass are then compared and discussed: the syngas biorefinery and the lignocellulosic biorefinery. Syngas biorefineries are less sensitive to biomass qualities and are technically relatively advanced, but require high investments and large-scale facilities to be economically feasible. Lignocellulosic biorefineries require multiple processing steps to separate the recalcitrant lignin from cellulose and hemicellulose and convert the intermediates into valuable products. The refining processes for high-quality lignin and hemicellulose fractions still need to be further developed. A concept of a modular lignocellulosic biorefinery is presented that could be flexibly adapted for a range of feedstock and products by combining appropriate technologies either at the same location or in a decentralized form.

KEYWORDS

biorefinery, lignocellulose, perennial crops, pretreatment, short rotation coppice, value chain

1 | INTRODUCTION

Lignocellulose is the term given to a type of biomass composed mainly of cellulose, hemicellulose and lignin (Carroll & Somerville,

2009). Trees, grasses and harvest residues from food crops are the major sources of lignocellulosic biomass. It is the most abundant biomass on Earth with an annual global production of about 181.5 billion tonnes (Paul & Dutta, 2018). Of these, about 7 billion tonnes

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from agricultural, grass or forest land are currently used as fodder or for energetic and material purposes (FAO, (2014); Piotrowski & Essel, 2015). Additionally, about 4.6 billion tonnes of lignocellulosic biomass residues are produced as agricultural residues, of which only about 25% are used intensively (Piotrowski, Essel, Carus, Dammer, & Engel, 2015). Traditionally, lignocellulosic biomass, especially wood, is used for heating and cooking, as a construction material and to produce fibres (e.g., in the pulp and paper industry). Recently, there has been increased interest in lignocellulosic biomass as a promising renewable resource for the bioeconomy (Kumar, 2014; Lewandowski, 2016). To meet the growing demand, optimized concepts to use the large potential of agricultural and forestry residues and improved sustainable cultivation systems for lignocellulose plants are required. Biorefinery concepts are being developed to convert lignocellulose into a broader spectrum of products and use all intermediates and sidestreams in optimized value networks by integrating various conversion and separation steps. Many of these activities are currently at pilot scale, such as the bioliq pilot plant in Karlsruhe (Germany) (Dahmen, Abeln, et al., 2016) and the lignocellulose biorefinery pilot plant in Leuna (East Germany), both of which are related to the Baden-Württemberg Research Network. The Leuna biorefinery uses the organosolv process as primary refining step. In addition, the pulp and paper industry has taken strategic initiatives to extend its pulp mills with the aim of becoming major players as biorefineries in the future bioeconomy by extending its portfolio from paper, packaging and tissue to a wide range of new products (RISE, 2015).

This special issue summarizes selected aspects of research on lignocellulose value chains using methodologies from multiple disciplines, including crop science, biotechnology and process engineering as well as socio-economics. Part of the work stems from a collaborative research network entitled “Lignocellulose as new resource platform for novel materials and products” funded by the German federal state

of Baden-Württemberg to develop key enabling technologies for the production and use of lignocellulosic biomass in sustainable value chains and their techno-economic and ecological assessment. The following sections discuss future perspectives for the sustainable supply of lignocellulosic biomass and the integration of pretreatment and biomass conversion options into modular lignocellulosic biorefineries.

2 | WHY USE LIGNOCELLULOSIC BIOMASS IN A GROWING BIOECONOMY?

Lignocellulosic biomass is the resource of choice for applications that benefit from its physical properties (e.g., wood for construction purposes) or chemical composition (e.g., paper production from cellulose). These products make use of the natural components and structures of lignocellulose. Lignocellulose mainly consists of natural polymers forming the cell wall, cellulose, hemicellulose and lignin (Figure 1). In addition, lignocellulosic biomass contains varying amounts of moisture, proteins, minerals and minor constituents, for example, resins in wood, depending on its origin.

In bioenergy applications, lignocellulosic biomass is currently used for heat and power production by combustion and, to a smaller extent, for biofuel production. However, the profitability of this sector very much depends on the fluctuating mineral oil prices and the subsidies so far granted. In addition, the expansion of bioenergy in Europe is hampered by the ongoing critical discussion on the sustainability of biomass supply (Lewandowski, 2015). In the longer term, the capacities for the production of heat and electrical power from other renewable resources, such as solar, wind and hydropower, can be extended through implementation of technological advances. It is expected that the

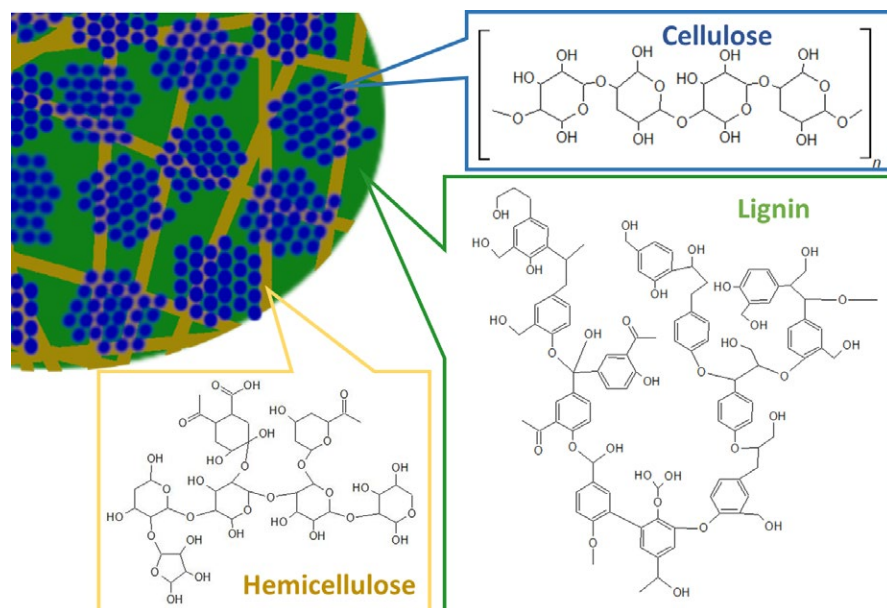


FIGURE 1 Cross section of a macrofibril of wood with the three major components of wood cell walls: cellulose (40–55 wt.%, linear C6 sugar glucose chains, polymerization degree 5,000–15,000, fibrils); hemicellulose (15–35 wt.%, branched C5 and C6 sugar chains, polymerization degree 100–1,000, amorphous); and lignin (20–40 wt.%, aromatic guaiacyl, coniferyl and syringyl alcohol monomers, three-dimensional network) (redrawn from a diagram by Sticklen, 2008)

use of lignocellulosic biomass for the production of biobased chemicals and materials will increase, since biomass is the only source of renewable carbon and due to its relative abundance and suitability. Against this backdrop, the EC recently decided to discontinue its support of the energetic use of forestry wood in the revised Renewable Energy Directive (RED) so as not to compete with the increasing demand for wood materials (www.paperage.com/2018news/01_17_2018cepi_redii.html). However, a certain expansion of bioenergy production capacities could still occur in biorefineries that integrate material and energy uses of biomass, but only use residual process biomass for energy purposes.

3 | WHAT ARE THE CURRENT STATUS OF AND FUTURE PERSPECTIVES FOR SUSTAINABLE BIOMASS SUPPLY?

The amount of lignocellulosic biomass that can be provided by good-practice forestry management is limited. For example, 232 million tonnes of wood was supplied in the EU 27 in 2011 (Piotrowski et al., 2015). Due to the limited potential of wood, the growing demand for lignocellulosic biomass will also need to be met by the agricultural sector, where it needs to be produced sustainably. Currently, about 3.7 billion tonnes of lignocellulosic biomass is supplied globally by grasslands, but mainly used as fodder. Another 1.3 billion tonnes come from agricultural residues and <1 billion tonnes from dedicated crops (Piotrowski et al., 2015).

In Europe, the discussion on competing biomass uses and future supply has led to the following criteria for “sustainable” biomass supply (see Lewandowski, 2015):

- The biomass is not used for food or animal feed purposes.
- The biomass is not needed to maintain ecological functions, such as soil humus content (agricultural production) or to replace nutrient withdrawal by harvested woods (forestry) or to support fauna (forestry).
- The biomass is grown on marginal land, which according to a definition by Elbersen et al. (2018) is land not suitable or economically attractive for food crop production.
- Where possible “regional” biomass is preferred, meaning that biomass is used in the same location as it is produced.

One of the aims of regional biomass supply is to reduce environmental impacts through its transport. Using the example of miscanthus, Wagner et al. (2019) demonstrated that transport distances up to 50 km only marginally affected the environmental performance of biobased products. In general, transportation and import of biomass should be carefully evaluated for its environmental, economic and social sustainability. For example, biomass production may secure income for rural populations. The associated risks such as the endangering of

smallholders’ land-use rights can be reduced through certification systems for sustainably produced biomass (Lewandowski & Faaij, 2006). Building biorefineries in countries with high biomass productivity, such as Africa and Latin America, and exporting processed biobased materials and intermediates could help to strengthen the economic situation in these areas and reduce environmental impacts through biomass transportation, provided market demand remains stable.

One factor favouring the use of lignocellulosic biomass that is often mentioned is the fact that it does not compete directly with food supply (Nanda, Azargohar, Dailai, & Kozinski, 2015). With regard to agricultural residues, alternative competing uses need to be taken into consideration. Many agricultural residues, especially cereal straw, have several other applications, such as animal bedding, and may at least partly be required to maintain humus content and soil fertility in intensively managed cropping systems (Blanco-Canqui & Lal, 2007; Memon et al., 2018). It is estimated that roughly 40% of agricultural residues need to remain on the field and another 20%–30% are diverted into various on-farm uses, mainly fodder (Daioglou, Stehfest, Wicke, Faaij, & van Vuuren, 2016).

A future, large potential for lignocellulosic biomass production is seen in the cultivation of dedicated perennial biomass crops (PBC) on agricultural land that is not needed or suitable for the cultivation of food crops (Dornburg et al., 2010; Hoogwijk, Faaij, & Eickhout, 2005; Smeets, Faaij, Lewandowski, & Turkenburg, 2007). However, this perspective has been criticized as it could lead to direct and indirect land-use change (ILUC). Direct land-use change occurs when one kind of land use replaces another, for example, when perennial biomass crops are established on cropland or replace forest or grasslands. GHG emission effects can be positive, for example, when perennial biomass crops replace annual crops and lead to carbon sequestration, or negative, when land-use forms with high carbon sequestration potential, such as forests, are replaced by biomass crops (Lewandowski, 2013). Indirect land-use change occurs, for example, when biofuel feedstock production triggers land-use change elsewhere due to the need to compensate for foregone food production on land now used for biofuels (HLPE, 2013). The calculation of ILUC effects is complex and requires establishment of the correlation between biofuel production in one place and new crop production established on former forest or grassland elsewhere. Modelling ILUC effects requires global scenarios (HLPE, 2013), which lead to high uncertainties and are not applicable for decision support on land use at regional or national level.

For the following reasons, we anticipate that PBC, in particular perennial grasses and the short rotation coppice (SRC) trees poplar and willow, will play an important role in the supply of sustainable regional lignocellulosic biomass to European biorefineries:

- PBC can be grown on marginal land and also have the potential to improve them. Marginal lands are often characterized

by biophysical constraints, including susceptibility to erosion, drought and salinity (Tóth, Montanarella, & Rusco, 2008). Perennial lignocellulose crops, such as miscanthus, switchgrass and poplar, are suitable for lands with such constraints because genotypes have been identified that are tolerant to abiotic stresses (e.g., drought, salinity, cold) frequently occurring on marginal land (Clifton-Brown et al., 2018; Lewandowski et al., 2016). However, Wagner et al. (2019) conclude that careful assessment of the specific prevailing conditions should be performed because biodiversity may be higher in some marginal lands under their existing vegetation than under PBC.

- PBC can have an environmentally beneficial performance. They only require soil cultivation once in a plantation lifetime of about 20 years, in the establishment phase. Conversion from annual to perennial cropping allows the soil organic carbon and associated properties to recover. The long-term soil rest reduces the risk of soil erosion and leads to soil carbon and humus accumulation, thus improving soil fertility and potentially improving degraded lands (Lewandowski, 2016).
- PBC can be integrated well into farming systems and provide them with additional biomass for on-farm use or as cash crops.
- PBC have the highest productivity of all biomass production systems in Europe. There are promising candidate PBC for temperate (see Clifton-Brown et al., 2018; Hoeber et al., 2018) and Mediterranean (see Fabbrini et al., 2018) European climates, including switchgrass, miscanthus, giant reed, willow and poplar. Breeding programs are in place for most relevant PBC. These exploit the genetic variability and have already delivered genotypes at varying levels of advancement (Clifton-Brown et al., 2018; Fabbrini et al., 2018; Hoeber et al., 2018)
- The chemical composition and physical properties of PBC biomass can be tailored to biomass utilization chains by the optimization of harvest time (Mangold, Lewandowski, & Kiesel, 2019a, 2019b), appropriate selection of available cultivars (Schäfer, Sattler, Iqbal, Lewandowski, & Bunzel, 2019) and, in the long term, through breeding programs (see Clifton-Brown et al., 2018). Biomass from PBC thus has a more consistent composition than biomass from wastes, which come from a variety of sources, and is therefore more suitable for conversion pathways with specific quality requirements.

4 | WHAT ARE PROMISING REFINING PATHWAYS FOR LIGNOCELLULOSIC BIOMASS IN FUTURE BIOREFINERIES?

Lignocellulosic biomass can be utilized in two main types of biorefineries: lignocellulosic and syngas biorefineries. In lignocellulosic biorefineries, the biomass is first separated

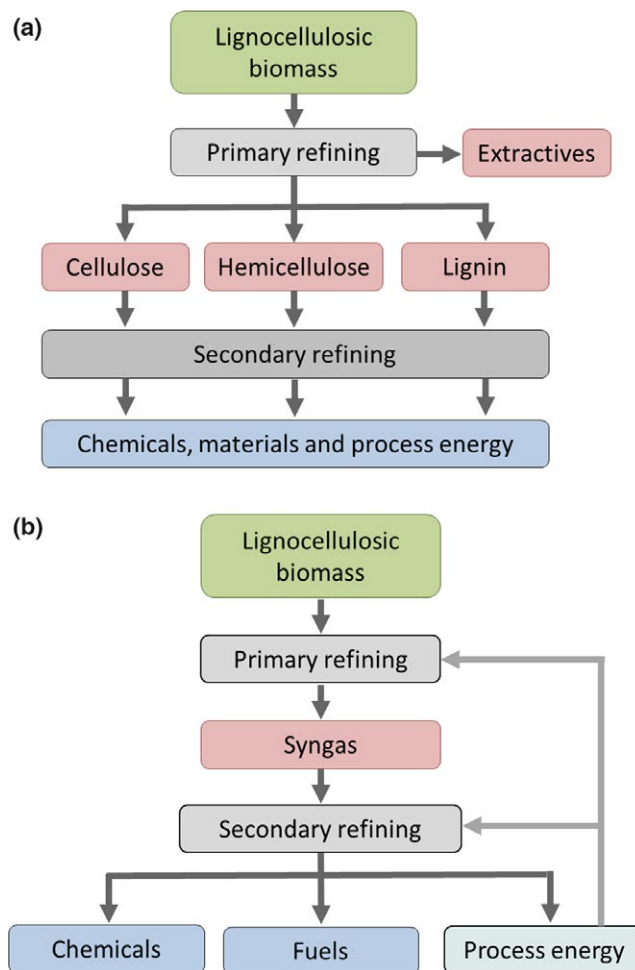


FIGURE 2 Biorefinery concepts making use of lignocellulosic biomass (adapted from BMBF (2012)). (a) The lignocellulosic biorefinery, characterized by: (i) Decomposition into natural intermediate units, for example, carbohydrates and aromatics; (ii) Lower conversion temperatures; (iii) Lower feedstock flexibility, parameters need to be adapted to the feedstock; (iv) A number of intermediates at the same time; (v) Products are platform molecules (C2–C4) from microbial and enzymatic conversion of sugars, monomeric, oligomeric and polymeric bio-aromatic fractions (>C6); (vi) Multistage process, parallel processing of biomass components; (vii) Can be built in modular form and close to the biomass source. (b) The syngas biorefinery, characterized by: (i) Complete decomposition of biomass into C1 units (CO) and hydrogen; (ii) High conversion temperatures; (iii) High feedstock flexibility; (iv) A single, defined intermediate after gas cleaning; (v) Products are hydrogen (for ammonia production), hydrocarbon fuels and bulk chemicals such as methanol and its derivatives. Heat and electricity are desired, unavoidable by-products; (vi) Complex high-temperature technology requiring large-scale operation for economic application; (vii) Requires infrastructure for long-distance biomass logistics; (viii) Process energy is an inevitable by-product by heat recovery from the high-temperature gasification process

into cellulose, hemicellulose and lignin (Figure 2a). These intermediates and the derived monomers constitute a versatile platform for further conversion into biobased chemicals

or biobased materials (Brodi, Vallejos, Tanase Opedal, Area, & Chinga-Charrasco, 2017; Harmsen & Hackmann, 2013; Lask, Wagner, Trindade, & Lewandowski, 2019). In syngas biorefineries (Figure 2b), lignocellulosic biomass is completely decomposed into synthesis gas (syngas) by the high temperatures applied in gasification processes (Dahmen, Henrich, & Henrich, 2017). After gas cleaning, the hydrogen and carbon monoxide produced can be processed into fuels and chemicals.

Even though the syngas and the lignocellulosic biorefinery compete for the same type of feedstock, they can be regarded as complementary approaches, in particular in terms of their feedstock and product portfolio. Figure 2 lists important characteristics of the two types of biorefinery.

The syngas biorefinery is much less sensitive to the type of feedstock due to the high-temperature treatment. Syngas, after gas cleaning, is a well-defined intermediate and can be converted into manifold products by so-called C1 chemistry, making use of technologies already established in the chemical industry that use coal or natural gas as feedstock. This way, synthetic hydrocarbon fuels by Fischer–Tropsch synthesis or platform chemicals, such as ethene and propene, can be produced via the intermediates methanol and dimethyl ether. It is expected that syngas biorefineries will only be economical at industrial scale with production capacities of several 100 kt/a due to the technical complexity and high temperatures of the processes involved. Syngas biorefineries can thus be considered as “centralized” plants, to which all biomass will have to be transported. Alternatively, biomass can be converted into an intermediate of higher energy density in decentralized plants before being supplied to a central large conversion facility. A comparative study for such concepts was performed within the EU FP7 project “BioBoost” for different thermochemical conversion pathways. The simulation tool developed for this purpose compared EU-wide biomass residue potentials, and various centralized and decentralized conversion technologies for fuel production. OpenStreetMap was used to raise realistic routing data (http://bioboost.eu/results/public_results.php). Syngas biorefineries have been developed up to technology readiness level (TRL) 7, depending on the specific technology used. In particular, syngas cleaning and chemical syntheses are already state of the art today based on technologies developed for coal and natural gas conversion processes. For this reason, syngas biorefineries could be implemented within a relatively short time frame in large scale. However, this technology today is considered to be only effective on large scale, demanding for significant investments and that the overall process still suffers from the insufficient market value of advanced biofuels.

The lignocellulosic biorefinery makes use of the molecular structures of all components contained in the biomass. Therefore, the type and quality of biomass plays an important role because it determines the pretreatment requirements

and primary refining steps to be applied and because certain molecular structures are desired and need to be preserved. Such a biorefinery is economically viable when each material stream is used in the value chain. Lignin could be used for the production of polymer materials and purified C6- and C5-sugar streams for fermentation processes. Economic assessment was done by Laure, Leschinsky, Fröhling, Schultmann, and Unkelbach (2014) on the basis of the conversion of 400,000 t/a dry wood using the organosolv processing. One case study showed that a competitive glucose price of 218 €/t could be achieved when a revenue of 325 €/t is obtained from the lignin and C5-sugar streams (Laure et al, 2014).

5 | WHICH CONCEPTS FOR LIGNOCELLULOSE BIOREFINERY ARE DISCUSSED?

A lignocellulosic biorefinery can be designed in a modular form, with the most efficient combination of different process modules being selected according to the available biomass, target products and production costs. As an example, out of four different constellations of an organosolv-based biorefinery it turned out that the combination of ethanol production along with recovery of food-grade CO₂ performed best. The yeast biomass from fermentation and the C5-sugar fractions were converted to biogas (Budzinski & Nietzsche, 2016).

In principle, lignocellulosic biorefineries may be realized at lower production capacities and could then benefit from lower logistics costs for biomass supply. It has been suggested that such small-scale biorefineries that can benefit from lower investment and transportation costs and increase circularity could play a future role, as example, as part of sugar refining plants (Kolfshoten, Bruins, & Sanders, 2014).

Lignocellulosic biorefineries could also be conceptualized as “on-farm biorefineries,” established close to the biomass production and run by farmers or cooperatives. An example of the practical integration of biomass production and conversion processes in decentralized lignocellulose value chains can be seen in on-farm biogas plants. Biogas is used on location for electrical power and heat production today. Alternatively, after clean-up and carbon dioxide separation, the methane can be fed into the natural gas grid. The digestates, which contain valuable plant nutrients, are brought back onto the field as organic fertilizer. In addition to energy production, there are several options for obtaining intermediates for materials and chemicals from the biogas value chain (see Bahrs & Angenendt, 2019). Thus, it would also be possible to incorporate a biogas plant as one module in a lignocellulosic biorefinery. Such a modular biorefinery integrating a biogas plant is being investigated by the EU project “GRACE” and a follow-up project to the

research network mentioned above. In this modular biorefinery concept, miscanthus is utilized to produce furfural and hydroxymethylfurfural from the sugar fraction as well as bio-aromatics from the lignin fraction in a small-scale plant.

Modular biorefinery concepts can link small-scale processing units to central large-scale units for further conversion steps. As an example, lignin could be supplied to large-scale conversion plants from a number of small-scale biorefineries, which only make use of the sugar fractions. The Horizon 2020 project AMBITION investigated scenarios where a number of 100,000 t/a pretreatments plants are combined to a larger gasification system, in which 200,000 t/a of by-produced lignin can be converted into syngas for use in fermentation.

Ultimately, it would be reasonable to integrate processes for the production of biobased chemicals from lignocellulosic biomass into existing biomass processing plants (e.g., sugar refineries or pulp and paper plants). This could generate value-added products and thus help to improve the economics of existing biomass processing plants. Already in the 19th century, a diversified chemical industry was established that made use of side products from wood and charcoal processing. However, due to the low economic performance compared to petrochemical products, only a few of these product pathways remain today, for example, vanillin from wood lignin and acetic acid from charcoal production.

The key technologies for lignocellulosic biorefineries are at very different phases of development. Flagship plants exist for second-generation bioethanol production along with a number of pilot-scale facilities (IEA, 2017). A few processes currently under development are at pilot scale (TRL 6), for example, the organosolv process; many others are still close to the proof-of-principle level. For that reason, significant R&D effort is required for lignocellulosic biorefineries,

not only for the technologies necessary for the individual processing modules, but also for the design of integrated biorefinery concepts that combine a selection of processing modules to establish site- and biomass-specific sustainable value networks that make use of essentially all components of lignocellulosic biomass.

The choice and combination of refining pathways and thus the design of a lignocellulosic biorefinery are steered by multiple factors: (a) the desired products, intermediates and sidestreams; (b) the amount and type of biomass and/or intermediates available; (c) the existing technology portfolio; and (d) appropriate conversion capacities of possible process modules.

6 | WHICH PROCESSES MAY BE CONTAINED IN A FUTURE LIGNOCELLULOSIC BIOREFINERY?

In the Baden-Württemberg Lignocellulose Research Network, biomass value chains were selected for development that have a ready supply of suitable feedstock and promising market opportunities in the region, along with appropriate refining technologies. The energetic use of sidestreams was included as an option for those fractions for which no other technology is available or economically feasible. Biomass sources from forestry and agriculture, including crops and residues, were considered, with a certain focus on miscanthus as the most productive energy crop in Baden-Württemberg, and poplar with bark as an example of short rotation coppice (SRC). The following sections discuss how the results obtained for the individual process modules can be potentially combined to form a future modular lignocellulosic biorefinery. In Figure 3, the different value chain options that were investigated are indicated

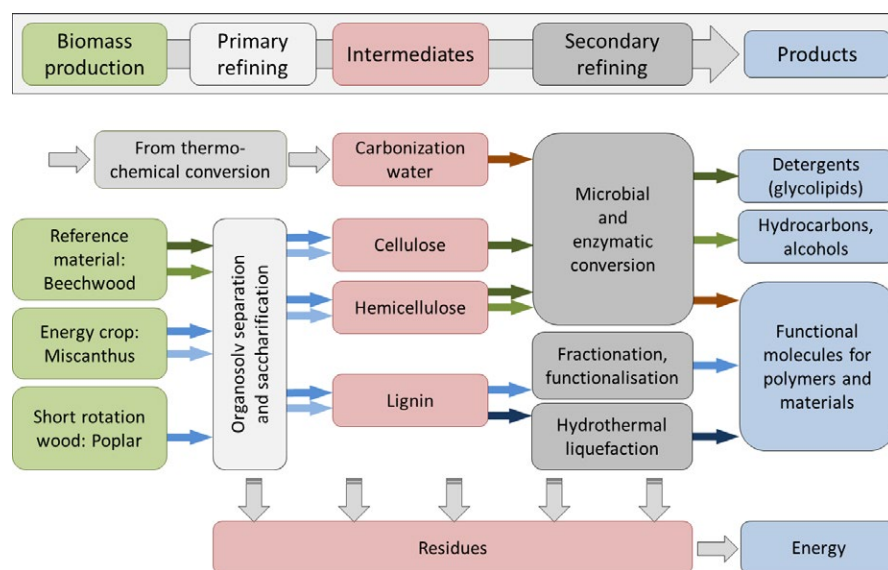


FIGURE 3 Value chains analysed in the modular lignocellulosic biorefinery concept of the Baden-Württemberg Lignocellulose Research Network. The coloured lines indicate selected value chains described in one of the research articles of this special issue: Seibert-Ludwig et al. (2018) (light blue), Rohde et al. (2019) (medium blue), Schuler et al. (2019) (dark blue), Siebenhaller et al. (2017), Horlamus et al. (2018), (dark green), Dörsam et al. (2016), Lange et al. (2017) (brown). Other data are taken from Lange et al. (2018) (medium green)

by coloured lines showing the path from primary refining via intermediates through to secondary refining and further conversion steps to different products.

For an optimal integration of biomass production and conversion, advanced breeding (see, e.g., Clifton-Brown et al., 2018) is required that tailors the biomass to user needs, resulting in improved pretreatment and conversion efficiencies. Taking miscanthus as an example (see column “Biomass production” in Figure 3), this can be achieved by selecting genotypes with a suitable cell wall composition (Schäfer et al., 2019) or high leaf share (Mangold et al., 2019b). Suitable agricultural practices can decrease the pretreatment requirements of lignocellulosic biomass, for example, by green harvesting PBC grasses and ensiling the biomass (Mangold et al., 2019a).

6.1 | Primary refining steps

Lignocellulose is a complex and relatively recalcitrant composite material. For this reason, several processing steps are necessary to completely utilize its components in a lignocellulosic biorefinery, where it is first separated into its natural components cellulose, hemicellulose and lignin (see column “Intermediates” in Figure 3). Typical mechanical pretreatment methods used to break down the relatively robust material are milling and grinding. This is followed by primary refining through a variety of possible methods. These include (among others) treatment with acid or base solvents (refer to column “Primary refining” in Figure 3), organic solvents such as organic acids, ketones and alcohols (e.g., acetone, methanol, ethanol) or ionic liquids to dissolve lignin from the fibres. Lignin can then be recovered by precipitation or by evaporation of the solvent. In the next step, the dissolved hemicellulose is recovered from the cellulose fibres (Zhang, Pei, & Wang, 2016). The study of Seibert-Ludwig, Hahn, Hirth, and Zibek (2019) systematically compared reaction conditions for separation processes applied to miscanthus and poplar wood. The aim was to find the most favourable process conditions to achieve a high grade of delignification and low cellulose solubilization, thus leading to a high availability of cellulose for enzymatic hydrolysis. The study compared alkaline, hot water, organic solvent as well as acid- and base-catalysed organosolv treatments. For the biomasses selected, it was found that acid-catalysed organosolv processing resulted in the highest delignification grade leading to a reasonably high glucose yield of above 70 wt.% yield after enzymatic saccharification for microbial conversion.

6.2 | Refining pathways for the use of lignin

Rohde et al. (2019) also applied the organosolv process (see column “Primary refining” in Figure 3) and subsequent thermal separation in order to obtain different lignin fractions suitable for chemical applications from miscanthus and

poplar. An industrial sulphonated lignin (Indulin AT) was used as standard reference. Low, medium and high molecular weight fractions were obtained by solvent extraction, successive precipitation and ultrafiltration. The most suitable separation method for organosolv lignin was found to be solvent extraction for poplar and successive precipitation for miscanthus, in terms of the best fraction properties for further chemical use (i.e., mass distribution, molar mass separation, polydispersity and functionality characterized by OH-group distribution). For the generation of chemical building blocks from lignin, fractionation is an important interim step, providing fractions of distinct structural and functional properties. High molecular lignin, for example, can be used in adhesives, carbon fibres and polymer blends (Wells, Kosa, & Ragauskas, 2013). The low molecular fractions generated are of particular interest for use in polymer synthesis and may serve as a bisphenol A substitute in the production of epoxy resins (Asada, Basnet, Otsuka, Sasaki, & Nakamura, 2015). In addition, the application spectrum of low molecular poplar lignin appears to be broader than miscanthus lignin due to its higher number of more reactive and sterically unhindered aliphatic groups. These results demonstrate the potentials of process optimization at a very early stage of the lignocellulosic value chain (Rohde et al., 2019).

After primary refining, the cellulose, hemicellulose and lignin fractions can be used in a variety of processes. As an aromatic polymer, lignin can be used in the development of adhesives and other biobased materials. Further decomposition of lignin molecules could provide monomeric and oligomeric aromatic compounds as candidates for building blocks in the chemical industry, but the efficient breakdown of lignin into chemical platform molecules has been the subject of research for many decades. Hydrothermal liquefaction (see column “Secondary refining” in Figure 3) of lignin appears to be a gentle method for this purpose (Toor, Rosendahl, & Rudolf, 2011). On the one hand, water is a natural solvent for biomass constituents and energy-intensive drying can be avoided. On the other hand, water acts as a reactant with higher selectivity than ethanol leading to higher catechol ($C_6H_4(OH)_2$) yields during solvolysis. As a bifunctional molecule, catechol is an interesting platform chemical for further conversion into polymeric materials. Today, about 20,000 tonnes are produced annually, mainly as a precursor for pesticides, flavourings and fragrances (ChEBI, 2018). It can be expected that a sustainable and economic production of catechol from biomass would lead to a dramatic increase in demand as these replace fossil-based chemicals. The same is true for many other biobased chemicals under development today. A kinetic model has been developed by Schuler, Hornung, Dahmen, and Sauer (2019) that predicts the product composition when hydrothermal liquefaction is applied to different types of biomass, at different reaction temperatures and with different reaction times. As with other lignin

depolymerization processes, hydrothermal liquefaction leads to several products. Proper analyses, separation and clean-up steps still need to be developed and integrated into the entire conversion process. For this purpose, expertise generated in the last century to produce aromatic compounds from tar, a by-product in the production of coke from coal, can be utilized. The main focus of current research is on conversion technologies, but in order to develop a complete process that allows for overall techno-economic assessment, research also needs to consider up- and downstream treatments.

6.3 | Refining pathways for the use of cellulose, hemicellulose and derived sugars

The cellulose and hemicellulose obtained can be hydrolysed to sugars and used as second-generation feedstock in various microbial and enzymatic processes. Some of these processes, for example, the production of biobased ethanol, lactic acid and succinic acid as building blocks for biobased polymers, have already been commercialized. A comprehensive overview of possible biotechnological products and metabolic pathways that make use of the cellulose-derived C6 sugars and the hemicellulose-derived C5 sugars is given in Straathof (2014). However, the organosolv hydrolysates are complex media, which may significantly influence microbial or enzymatic syntheses. Thus, the efficient use of substrate mixtures as well as the sensitivity of the biologic systems to side products and inhibiting components is an important aspect in process development. Such effects are being investigated by a number of projects in the Baden-Württemberg Lignocellulose Research Network. Siebenhaller et al. (2017) have developed a lipase-catalysed method of producing glycolipids (rhamnolipids) from glucose- and xylose-rich sugar mixtures derived from beechwood via an acid-catalysed organosolv process followed by hydrolysis (see column "Secondary refining" in Figure 3). This new method involves the utilization of a deep eutectic solvent and is therefore an elegant way of overcoming the low solubility of sugars in other water-free solvents. This opens up interesting perspectives for the future use of lignocellulosic biomass but requires further optimization to increase yields.

Hemicellulose is a polymer made up of long chains of various sugar molecules, including a high proportion of C5 sugars, which cannot be efficiently used as a substrate by most microorganisms. For this reason, dedicated work has been conducted in the Lignocellulose Research Network to modify microbial strains by means of metabolic engineering with suitable enzymes enabling them to grow on C5 sugars. Work by Lange, Müller, Takors, and Blombach (2018) has enabled *Corynebacterium glutamicum* to produce isobutanol in an anaerobic two-phase process utilizing a hemicellulose fraction obtained from beechwood organosolv processing.

6.4 | Valorizing syngas biorefinery sidestreams

Further projects in the research network aimed at valorizing sidestreams of the syngas biorefinery using fermentation approaches. The carbonization water derived from fast pyrolysis as the primary refining step in the bioliq biorefinery (Dahmen, Pfitzer, et al., 2016) contains up to 30 wt.% of dissolved organic substances, which should not be treated as waste, but used as feedstock for further conversion (carbonization water in Figure 3). Microbial use and conversion of this process water have been investigated using fungal (*Aspergillus oryzae*), gram-positive (*Corynebacterium glutamicum*) and gram-negative (*Pseudomonas putida*) bacterial production systems; however, growth-inhibiting components in the carbonization water lead to low tolerance levels for these microorganisms. A systematic study using model substances (including aldehydes, organic acids and phenolic substances) represented in the carbonization water led to the determination of maximum concentrations allowing growth and organic acid (malate) production by *Aspergillus oryzae* (Dörsam et al., 2016). In other work, protocols for the pretreatment of the carbonization water have been established that enable conversion of its major constituents, acetate and acetol, into 1,2-propanediol (Lange et al., 2017). These examples serve as evidence that pretreatment of carbonization water is required prior to fermentation. As with other fermentation processes, downstream processes for separation and product cleaning also need to be considered in the development of biorefineries.

The refining pathways presented here can only be regarded as exemplary modules of a biorefinery. For biorefineries composed of these modules, life cycle and techno-economic assessment still need to be conducted to identify favourable refining pathway constellations in terms of ecological, economic and carbon efficiency. This will require data on material and energy balances as well as further information of the processes and products involved.

To convert concepts into practice, projects have been launched in a second round of funding within the Baden-Württemberg Bioeconomy Research Program in close cooperation with relevant stakeholders from industry to identify useful biobased products.

7 | CONCLUSION— PERSPECTIVES FOR INTEGRATED LIGNOCELLULOSIC VALUE CHAINS

The supply of sustainably produced biomass is the prerequisite for a lignocellulosic value chain to perform well. It can be provided from sustainable forestry and from agricultural production,

with agricultural residues and perennial biomass crops (PBC) being the most favourable resources (Clifton-Brown et al., 2018; Fabbri et al., 2018; Hoeber et al., 2018). PBC can be integrated into existing farming systems using less favourable land or land marginal for food crop production, with the additional provision of various ecological benefits (Wagner et al., 2019).

The decisive factors in a viable overall lignocellulose biorefinery concept will be the biomass feedstock potential and supply, the technology platform available, and—most important—the product demand and value. From the studies described and discussed here, it becomes clear that modular lignocellulosic biorefinery concepts have several advantages: The combination of units that perform individual refining steps of the value chain helps to design biorefineries tailored for specific locations, products and markets. Using similar units in several places will reduce development and investment costs and therefore make small-scale biorefineries more feasible. In the light of rapid technology development and changing market demands, modular thinking can also prepare for easier adaptation of the process chains.

The definition of the appropriate scale of conversion capacity remains as important question. Price supply curves for biomass production and logistics, economy of scale for installations costs of refining plants, as well as product yield and value will mainly determine the economics and thus the reasonable size of a lignocellulosic biorefinery.

The longer term implementation of the biorefineries will lead to a transition of the resource platform from fossil-based to renewable resources. Initially, fully compatible biobased “drop-in” products will be phased into the otherwise mostly fossil-based product world. This can be achieved largely using existing processes and others adapted to biomass as feedstock instead of fossil fuels. Then the value chain and product portfolio flexibility will be increased through the gradual integration of new chemical and biochemical processes. Consequently, this will result in a new chemical and technical platform based on lignocellulosic biomass. The transition to this new platform and the development of appropriate technologies will take time. However, the development of crude oil refineries also took several decades before the highly integrated and optimized facilities we know today were in place. Likewise, biorefineries will start with a limited number of products and expand over time with the increasing degree of integration and diversification and the development of new process modules. In analogy to crude oil refineries, no two of which are identical, different biorefinery configurations will be realized according to the business model applied by their owners, the markets to be served and the feedstock utilized.

Both syngas and lignocellulosic biorefineries, along with other types of biorefinery, will play a role in the utilization of lignocellulosic feedstock. However, the different parts of process chains under development today are usually developed independently from each other. Consistent work on full process chains is still rare, particularly at TRL levels above 4. Research

and development activities increasingly need to address process integration along the value chain. This includes additional pre-treatment or tailoring of biomass, separation, conditioning and upgrading of intermediates prior to further use and enhanced valorization of the products. For process integration, energy and mass flows need to be optimized along the entire value chain (Budzianowski & Postawa, 2016; Nikolakopoulos & Kokossis, 2017). First and foremost, the product portfolio and how it fits into existing and possible future markets needs to be considered. Today, biorefinery development is strongly feedstock and technology-driven, but not so much from the product demand side. Dedicated tools to support the implementation of innovative technologies and prepare the market uptake of new products will be needed to accelerate the transition.

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